



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Yeats, K. et al
Serial No. : 09/759,031
Filed : January 12, 2001
For : Ambient Temperature Curing Coating Composition
Group Art Unit : 1712
Examiner : Moore, Margaret G.
Docket No. : ACO 2758US

DECLARATION UNDER 37 CFR 1.132

I Keith Yeats do hereby declare as follows:

- 1) I am a citizen of the United Kingdom residing at 26 Redford Place, Burradon, Cramlington, Northumberland, NE23 7LG, UNITED KINGDOM.
- 2) I am a Research Associate at International Coatings (an Akzo Nobel company), where I have worked for 22 years. I attended Sheffield City Polytechnic from 1977-1980, where I obtained a Higher National Diploma in Chemistry. Between 1982-1986, I attended Newcastle Upon Tyne Polytechnic where I achieved a First Class Honours degree in Applied Chemistry (BSc Hons). I went on to obtain a Ph.D. in Chemistry at the University of Durham in 1990.
- 3) I conducted and/or had control over the following experiments (all quantities are given as parts by weight)

Preparation Of Acrylic Polymer

- i. An acrylic polymer was prepared in a reaction vessel equipped with a mechanical stirrer, nitrogen inlet tube, temperature controller, reflux

condenser and inlet tubes for the addition of monomers. Dow Corning DC 3074 (69.33 parts) was charged to the vessel and heated to 140°C with stirring under nitrogen. A mixture of butyl acrylate (17.08 parts), glycidyl methacrylate (12.63 parts) and Trigonox B (0.98 parts) was added over a period of 4 hours. After the addition was completed, the temperature was maintained at 140°C for 2 hours. The product was allowed to cool before being discharged from the reaction vessel.

- ii. An acrylic polymer was prepared using the same process as in i) except that Silres SY231 (69.33 parts) was used in place of the DC 3074.
- iii. A comparative acrylic polymer was prepared. Trimethylbenzene (24.27 parts) was charged to the reactor and heated to 140°C with stirring under nitrogen. A mixture of butyl acrylate (42.16 parts), glycidyl methacrylate (31.17 parts) and Trigonox B (2.41 parts) was added over a period of 4 hours. After the addition was completed, the temperature was maintained at 140°C for 2 hours. The product was allowed to cool before being discharged from the reaction vessel.

Note:

DC 3074 and Silres SY 231 are reactive diluents in accordance with the claimed invention. They each have a viscosity of less than 2 Pa.s (20 Poise) at 25°C and have at least one functional group which is substantially non-reactive with olefinically unsaturated monomers and which is capable of reacting with a curing agent to form a polymer network.

DC 3074 is described by Dow Corning as being a methoxy terminated dimethyl phenylsiloxane, with the following typical properties: Molecular weight 1000-1500, phenyl/methyl ratio 1.3/1.0, Si-OCH₃ content 15-18%.

Silres SY 231 is described by Wacker as being a methoxy-functional methyl phenyl siloxane. Analysis carried out by International Coatings found an approximated molecular weight (Mn) of 1000. The approximate mole ratio of organic functional groups was phenyl: methyl: methoxy: butoxy 33: 39: 23: 5.

Trimethylbenzene, the diluent utilized in preparing the comparative polymer (iii), is not capable of reacting with a curing agent to form a polymer network.

Preparation Of Coating Composition

The following examples of coating compositions made in accordance with the claimed invention include one of the polymers (i) or (ii) above which are made by polymerisation in the presence of a reactive diluent as claimed. The comparative coating compositions include comparison polymer (iii) above which is made by polymerisation in the presence of a diluent not in accordance with the claimed invention. However, a reactive diluent in accordance with the claimed invention is mixed in during preparation of the comparative coating compositions.

- iv. A pigmented coating composition was prepared: Polymer from i) (58.45 parts), titanium dioxide (26.25 parts), trimethylbenzene (7.0 parts), Dysperbyk 111 (0.79 parts), Byk 080 (0.21 parts), Modaflow resin (0.10 parts) and Thixatrol ST (1.60 parts) were stirred together using a high-speed stirrer. A curing agent (3-aminopropyltriethoxysilane, 5.32 parts) and a catalyst (dibutyltin diacetate, 0.28 parts) were added and the mixture was stirred by hand immediately before applying to test panels.

A pigmented film A was prepared by applying the coating to aluminium panels using a 200-micrometer wire-wound applicator. The coated panels were left for 24 hours at room temperature before a second coat was applied. Then, the coated panels were allowed to dry for 7 days before testing the gloss retention in accelerated weathering tests.

- v. A comparative pigmented coating composition was prepared: Polymer from iii). (24.08 parts), Titanium dioxide (26.65 parts), DC 3074 (39.85 parts) Dysperbyk 111 (0.79 parts), Byk 080 (0.21 parts), Modaflow resin (0.10 parts) and Thixatrol ST were stirred together using a high-speed stirrer. A curing agent (3-aminopropyltriethoxysilane, 5.29 parts) and a catalyst (dibutyltin diacetate, 0.28 parts) were added and the mixture was stirred by hand immediately before applying to test panels.

A pigmented film B was prepared by applying the coating to aluminium panels using a 200-micrometer wire-wound applicator. The coated panels were left for 24 hours at room temperature before a second coat was applied. Then, the coated panels were allowed to dry for 7 days before testing the gloss retention in accelerated weathering tests.

- vi. An unpigmented coating composition was prepared: Polymer from i) (41.1 parts) trimethylbenzene (4.9 parts) 3-aminopropyltriethoxysilane, (3.7 parts) and dibutyltin diacetate (0.2 parts) were stirred together by hand immediately before applying to glass test panels.

An unpigmented film C was prepared by applying the coating to glass panels using a 300-micrometer draw down bar. The coated panels were stored in an incubator at 25 °C. The development of the hardness of the film was determined by applying a König hardness test to panels that were temporarily taken out of the incubator.

- vii. An unpigmented coating composition was prepared using the same formulation as in vi) but replacing the polymer from i) with the polymer from ii) (41.1 parts).

An unpigmented film D was prepared by applying the coating to glass panels using a 300-micrometer draw down bar. The coated panels were

stored in an incubator at 25 °C. The development of the hardness of the film was determined by applying a König hardness test to panels that were temporarily taken out of the incubator.

- viii. A comparative unpigmented coating composition was prepared: Polymer from iii) (16.7 parts), DC 3074 (28.5 parts), trimethylbenzene (0.9 parts), 3-aminopropyltriethoxysilane, (3.7 parts) and dibutyltin diacetate (0.2 parts) were stirred together by hand immediately before applying to glass test panels.

An unpigmented film E was prepared by applying the coating to glass panels using a 300-micrometer draw down bar. The coated panels were stored in an incubator at 25 °C. The development of the hardness of the film was determined by applying a König hardness test to panels that were temporarily taken out of the incubator.

- ix. A comparative unpigmented coating composition was prepared using the same formulation as in viii) except that Silres SY 231 (28.5 parts) was used in place of DC 3074.

An unpigmented film F was prepared by applying the coating to glass panels using a 300-micrometer draw down bar. The coated panels were stored in an incubator at 25 °C. The development of the hardness of the film was determined by applying a König hardness test to panels that were temporarily taken out of the incubator.

The experiments were formulated so that tests could be carried out on films having the same overall composition, so that the effects of the different methods of polymer synthesis could be compared.

4) The test results of the experiments were as follows

Gloss retention tests

The pigmented films A and B were exposed to accelerated weathering tests in QUV (A) cabinets. The gloss retention (% non-scattered light) of the films was measured, see Table 1.

Table 1 Gloss retention after exposure to UV(A) light

Exposure time	0 hours	49 hours	217 hours	648 hours	986 hours
	Non-scattered light (%)	Non-scattered light (%)	Non-scattered light (%)	Non-scattered light (%)	Non-scattered light (%)
Coating according to the current invention (Pigmented film A)	100	100	100	99	99
Comparative coating (Pigmented film B)	100	85	79	78	78

After exposure to UV(A) light, the gloss retention of the pigmented coating prepared according to the current invention was higher than the gloss retention of the comparative pigmented coating.

Film hardness tests

The film hardness of the unpigmented films C, D, E, and F was determined using the König hardness test. This involves a pendulum which swings while being supported by two ball bearings which rest on the coating. The higher the number of swings before the pendulum stops, the harder the coating is. The test results are presented in Table 2.

Table 2 Development of film hardness after application

Time after application	1 week	2 weeks	4 weeks
	Hardness (number of swings)	Hardness (number of swings)	Hardness (number of swings)
Coating composition according to the current invention, using DC 3074. (Unpigmented film C)	64	80	93
Coating composition according to the current invention, using SY 231. (Unpigmented film D)	50	71	85
Comparative coating composition using DC 3074 (Unpigmented film E)	39	44	53
Comparative coating composition using SY 231 (Unpigmented film F)	29	33	40

The development of the film hardness of the coating composition according to the current invention was faster than the development of the hardness of the comparative coating composition. This can be seen in Table 2 by comparing the results for C against E and comparing the results for D against F.

5) Conclusions

The coating compositions comprising a glycidyl-functional acrylic polymer obtained by polymerisation in the presence of a reactive diluent, in this case

polysiloxane DC 3074 or polysiloxane Silres SY 231, resulted in coatings having superior gloss retention and hardness properties compared to coatings prepared from compositions obtained by mixing the components.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent resulting therefrom.

DATE: 27th November 2002

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